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# Upper Stratospheric (30–50 km) Lidar Observations of the Ozone Vertical Distribution

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A maximum reduction of 20% of the ozone content in the altitude range 35–45 km is presently expected within the next 50 years. Early detection of trends is thus of particular importance to confirm such extrapolations. An active lidar system, which uses a powerful exciplex laser as the emitting source at 308 nm, has been operated at the Observatoire de Haute Provence to probe the upper stratospheric ozone distribution. A description of the lidar system and results of the measurements are given and compared with those obtained simultaneously by already proven techniques such as Brewer-Mast sondes and Umkehr spectroscopic measurements. A general agreement within the uncertainties of the various methods is found. The data are further analyzed to determine a total error budget and to extrapolate the potentiality of lidar systems, using up-to-date laser sources to reach a precision better than 3% in the considered altitude range.

## 1. INTRODUCTION

Anthropogenic emissions of nitrogen and chlorine source species ( $\text{N}_2\text{O}$ , chlorofluoromethanes) have been shown to represent a potential threat to the atmospheric ozone layer, leading to modifications in the radiative and chemical balance of the earth's environment. Whereas at the present time the calculated total content variation in the ozone column stays below 5%, large modifications of the ozone vertical distribution are expected to occur [National Academy of Sciences, 1982]: at altitudes between 35 and 45 km a maximum reduction of 20% is presently extrapolated as a result of chlorine-activated destruction cycles. On the other hand, an increase of  $\sim 20\%$  is expected below 15 km because of  $\text{NO}_x$  emissions. Because of the radiative properties of ozone, both in the UV and IR wavelength ranges, such variations will greatly modify the thermal budget of the coupled troposphere-stratosphere system and directly influence the earth's climate. Detection of ozone trends is thus of particular importance to confirm the present day theories; it presently relies on both ground-based (Dobson spectrophotometer using the Umkehr technique) and satellite-borne passive experiments such as backscattered ultraviolet and solar backscattered ultraviolet experiments [World Meteorological Organization (WMO) 1982]. Such systems, however, have been recently shown to have intrinsic limitations, mainly due to calibration problems inherent to passive sensors [Fleig *et al.*, 1980].

Consequently, during the last few years, active lidar profiling of the ozone vertical distribution by the differential absorption laser (DIAL) technique in the UV wavelength range has been developed using two different type of laser sources: (1)  $\text{Nd}^{3+}$ : Yag pumped dye lasers which enable a large tuning range of the UV-emitted wavelengths from 280 to 305 nm, thus allowing measurements both in the troposphere and stratosphere up to altitude levels between 35 and 40 km [Pelon and Mégie, 1982a, b]; and (2) exciplex laser sources using xenon chloride ( $\text{XeCl}$ ) as an active medium and emitting at 308 nm, the off-wavelength being usually generated by Raman-shifting techniques [Uchino *et al.*, 1983; Werner *et al.*, 1983].

Such systems have proven their ability to study the ozone number density variations in the troposphere and stratosphere associated with mesoscale dynamic processes [Pelon and Mégie, 1985] and short-scale processes taking place within tropopause fold events occurring in the mid-latitude regions [Vernin and Pelon, 1985]. However, the rapid decrease both in atmospheric total density, which provides the support for light backscattering, and in ozone number density, which relates to the local optical thickness to be measured, makes measurements above 35 km very difficult without greatly increasing the laser-emitted power. The purpose of this paper is to describe the potentiality of lidar systems using a powerful exciplex laser source as the emitter to actively probe the upper stratospheric ozone distribution (30–50 km). Following the description of the experimental setup and related parameters, results of a first experiment will be presented, which will be used as a comprehensive basis for further extrapolation of the system accuracy with respect to the early detection of ozone concentration trends density in the upper stratosphere.

## 2. EXPERIMENTAL SYSTEM

The lidar system used for ozone measurements at the Observatoire de Haute Provence ( $44^\circ\text{N}, 5^\circ\text{E}$ ) since 1981, and the principle of the DIAL method have already been presented in detail by Pelon and Mégie [1982a]. In September 1983 an exciplex laser source was added to the system to probe the ozone vertical distribution in the upper atmosphere. The characteristics of the experimental setup are given in Table 1.

The emitter was an  $\text{XeCl}$  exciplex laser provided by the SOPRA Company under contract with the Centre National d'Etudes Spatiales (CNES) in France. At the time of the experiment this system was in its first phase of development, and its emitting characteristics were therefore limited as far as the emitted energy is concerned. It included a single-oscillator cavity in a two-plane mirror configuration. The active medium was a gas mixture of  $\text{Ar}/\text{He}/\text{Xe}/\text{HCl}$ . Because of the large divergence of the system in this multimode configuration (5 mrad), the output energy had to be reduced from its nominal value (200 mJ) by spatial filtering to a value of 70 mJ, compatible with a 1-mrad maximum divergence adapted to the lidar receiver field of view. The pulse repetition rate was 20 Hz. No spectral selection was made in the oscillator cavity, so that the emitted spectrum included three laser lines, at 307.71, 307.96,

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TABLE 1. Characteristics of Lidar System Used at the Observatoire de Haute Provence in September 1985

	Parameter	Value
Emitted energy	$E_1$	70 mJ (308 nm)
	$E_2$	60 mJ (355 nm)
Repetition rate	$r_1$	20 Hz (308 nm)
	$r_2$	10 Hz (355 nm)
Telescope area		$0.5 \text{ m}^2$
Optical yield	$\eta_1$	6% (308 nm)
	$\eta_2$	3% (355 nm)

and 308.21 nm, as recorded by a high-resolution spectrometer (Figure 1). The relative energy emitted in each of these lines had been measured and corresponded respectively to 2, 63, and 35%. Having characterized the laser spectral emission, it was then possible to compute the effective integrated absorption cross section of ozone over the laser lines; the resulting value was  $1.20 \times 10^{-19} \text{ cm}^2$  at 243 K. The absolute ozone cross-section values in this wavelength interval (307–309 nm) in the Huggins bands were taken from the recent results of Bass and Paur [1985] and correspond to a total relative variation over the laser-emitted spectral range of less than 3% (Figure 1) (see section 4 for a discussion of the related error on the ozone determination).

The off wavelength required for the DIAL measurement corresponded to the third harmonic of a  $\text{Nd}^{3+}$ : Yag laser (355 nm) already available at the lidar facility. The wavelength interval of 47 nm between the two emitted wavelengths is very similar to the one obtained by Raman shifting in an hydrogen cell [Loree et al., 1979]. Its large value precluded any measurement at an altitude level below 25 km, since during September 1983 the aerosol load in the stratosphere following the March 1982 eruption of El Chichon was still very high. The off wavelength of the DIAL measurement (355 nm) was thus used to monitor simultaneously the atmospheric backscattering coefficient between 15 and 50 km.

Two types of information can then be derived from this measurement: (1) The aerosol scattering ratio at 355 nm can be determined by comparing the lidar data with the atmospheric density profile measured by a meteorological rawinsonde at the same location. Scattering ratios up to 1.6 have

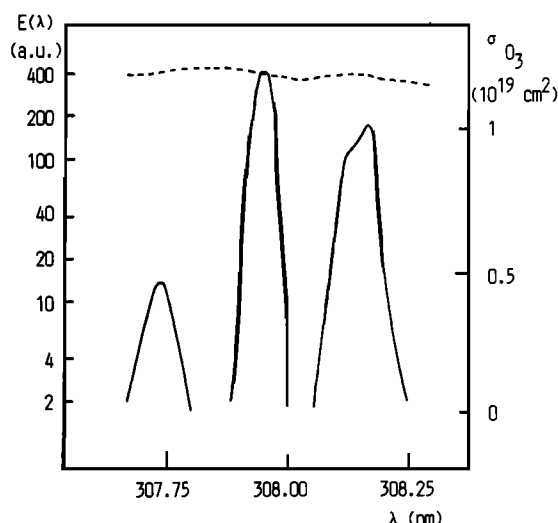


Fig. 1. Recorded spectrum of the emitted XeCl laser impulsion (solid line) as compared to the variation of the ozone absorption cross section near 308 nm (dotted line).

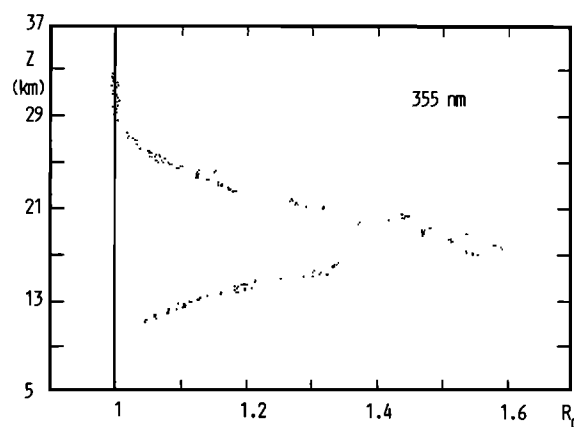


Fig. 2. Typical aerosol-scattering ratio recorded by lidar sounding at 355 nm in mid-September 1983 at the Observatoire de Haute Provence.

been observed around 20 km, whereas no aerosol layer was detected above 30 km (Figure 2). This result was further confirmed by lidar observations performed at 532 nm during the same time period. (2) Assuming thus that pure molecular scattering is only present above 30 km, the atmospheric density profile up to 50 km is determined by normalizing the lidar signal to the density values obtained from the rawinsonde data between 30 and 35 km. This allows the calculation of the Rayleigh extinction used to correct the ozone concentration profile [Pelon and Mégie, 1982a] and the determination of the ozone mixing ratio distribution, which will be used for further comparisons with in situ measurements (see section 3). Finally, the temperature profile in the same altitude range can be determined, assuming hydrostatic equilibrium and a pressure boundary condition [McCormick et al., 1967; Hauchecorne et al., 1980].

For the high-altitude measurements considered here, the backscattered signals are time sampled at 0.5 MHz, using the photon-counting mode. The data reduction used to derive the ozone number density is similar to the one described by Pelon and Mégie [1982a].

### 3. RESULTS

The above-described system was operated during 5 nights in September 1983 at the Observatoire de Haute Provence. These measurements were performed within the framework of the Middle Atmosphere Program (MAP) Global Budget of Stratospheric Trace Constituents (GLOBUS) International Campaign [Offermann, 1984], allowing comparisons with other measuring techniques.

Figure 3 shows the average profile of the ozone distribution between 25 km (above the ozone maximum) and 48 km, integrated over 5 nights between September 17 and September 24, 1983. The total integration time of 4 hours corresponds to  $10^5$  laser shots sequentially emitted on each wavelength. The altitude resolution is determined by the smoothing filter applied to the rough data and ranges from 0.6 km at the lower altitudes to 7.2 km at the uppermost level. The error bars plotted on this profile correspond only to the statistical error computed from the variances of the individual signals [Pelon and Mégie, 1982a]. Up to 30 km this error stays below 2%; it increases rapidly above this altitude and reaches 18% at 45 km. Figure 4 shows the ozone mixing ratio distribution determined from the lidar-measured atmospheric density profile, as indicated in section 2.

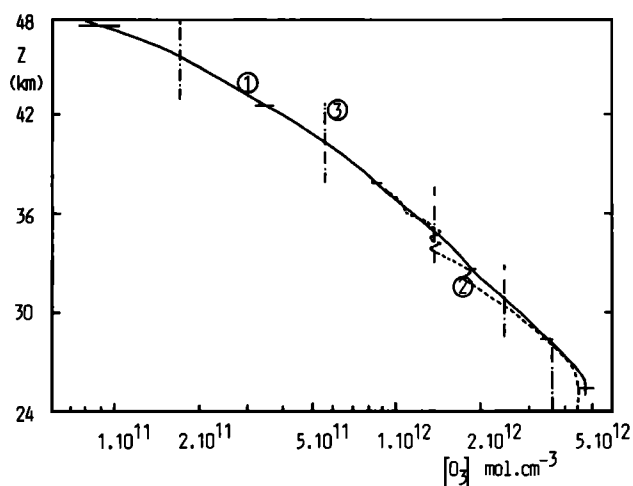


Fig. 3. Lidar averaged ozone profile (curve 1, solid line) during 5 nights between September 17 and September 24, 1983, compared to the average balloon-borne Brewer-Mast sondes profile (curve 2, dotted line) and to the average Umkehr profile (curve 3, vertical dotted line) obtained during the same period of time.

During the course of the MAP-GLOBUS campaign several other techniques were used to measure the ozone vertical distribution, including ground-based UV, IR, and microwave spectrophotometers and balloon-borne ozonsondes. At the Observatoire de Haute Provence, two systems were operated routinely: the Dobson spectrophotometer (number 85) automated for Umkehr measurements [Komhyr *et al.*, 1985] and balloon-borne Brewer-Mast ozonsondes. A comparison of the results of these two instruments (averaged between September 17 and September 24) with the lidar measurements is given in Figure 3. At lower altitudes the sonde data can still be considered as reliable, whereas some scattering in the measurements above 30 km can already be detected due to pump efficiency problems. Below 30 km, as already demonstrated in the case of the dye laser emitter [Pelon and Mégie, 1982b], good agreement is obtained within the accuracies of the two instruments. Considering the Umkehr data, the conversion from partial pressure to ozone number density has been made using a combination of radiosonde data and lidar data to calculate the vertical temperature distribution, as described previously. The temperature effectively used in the computation is then the temperature measured at the pressure level corresponding to the mean altitude of the Umkehr layer. As represented in Figure 3, taking into account the difference in altitude resolution of the two systems, the comparison leads again to a rather good agreement. The differences for layers 7 to 9 (32 to 48 km) are lower than 3%, for example, within the error bars of the measurements, including the uncertainties due to the calculation of the ozone concentration from Umkehr measurements and the accuracy on the ozone absorption cross sections. In layers 5 (23.5 to 28 km) and 6 (28 to 32 km), Umkehr values are lower by 20 and 8% respectively. This difference can most likely be attributed to the presence of aerosols at lower altitudes (cf. Figure 2) [DeLuisi, 1979]. Whereas these measurements already give a fair evaluation of the lidar system accuracy for ozone measurements above 40 km, a definitive answer with respect to the potentiality of this system for ozone trends monitoring will be given only after completion of numerous measurements allowing a statistical analysis of the system performances. However, these preliminary measurements can be used to evaluate the measurement

precision which will be obtained with up-to-date exciplex lidars as presently achievable, taking into account both statistical and systematic errors.

#### 4. DISCUSSION

As pointed out previously, one of the major goals in the monitoring of the stratosphere is the early detection of any ozone depletion at the 40-km altitude level. In this respect the main advantage of an active system such as the lidar is the operator control of the source, which leads to autocalibration of each system at any time and thus its independence within a given network. This is due to the fact that the ozone number density is directly derived by ratioing the signals themselves for two altitude levels and two wavelengths each. The only assumption to be made is then the constant time response of the system, in microseconds range, which corresponds to the height resolution.

Two types of errors have to be considered, as already analyzed by Pelon and Mégie [1982a]: (1) the statistical error related to the signal to noise ratio and to the local ozone absorption to be measured (this error is dependent on the choice of the emitted wavelengths and on the system parameters), and (2) systematic errors related to the accuracy of the measured effective ozone absorption cross section and its temperature dependence as well as to potential interferent extinction and absorption processes.

The optimization of range-resolved DIAL measurements in terms of operating wavelength depends in principle on three different parameters [Mégie and Menzies, 1980]: (1) the efficiency  $\rho$  of the scattering process, which varies as  $\lambda^{-4}$  if one considers Rayleigh scattering to be the dominant process, (2) the integrated on-line optical thickness  $\tau$  between the emitter and the scattered volume, and (3) the local optical thickness  $\Delta\tau$ , to be measured within the height resolution of the lidar system. Neglecting temperature variations of the absorption cross section with height,  $\Delta\tau$  can be considered to the first order, as far as wavelength dependence is concerned, as directly proportionnal to  $\tau$ .

Over a 20-nm wavelength interval between 290 and 310 nm,  $\rho$  only varies by 25%, whereas the variation of  $\tau$ , which is directly related to the wavelength dependence of the ozone

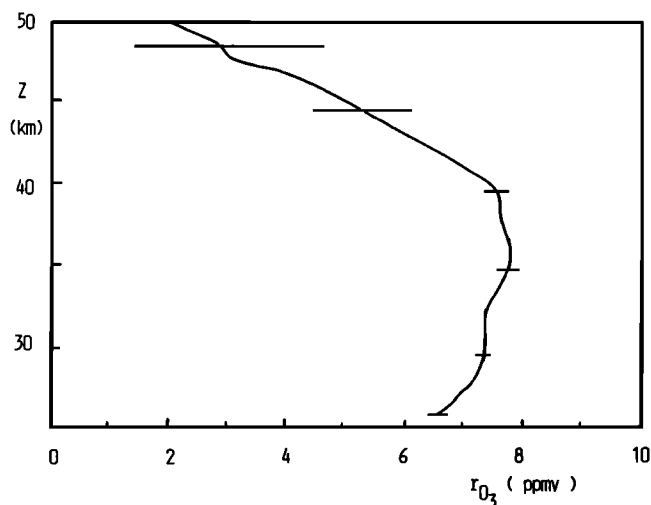


Fig. 4. Same lidar average ozone profile as Figure 3 but calculated in terms of mixing ratio, using the atmospheric density values deduced from the off-wavelength measurements.

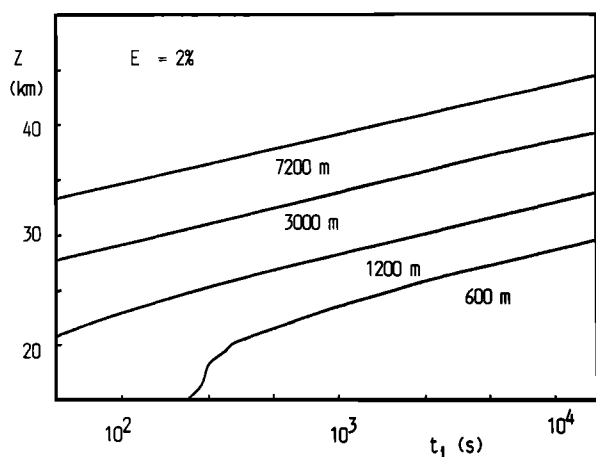


Fig. 5. Integration time  $t_i$  necessary to achieve a 2% accuracy for the lidar ozone measurements as a function of height for different height resolutions (simultaneous emission and detection at both wavelengths).

absorption, varies by a factor of 12. The integrated optical thickness is thus the relevant parameter to be optimized by the choice of the operating on-line wavelength; its optimum value is then shown to be 1.1 [Mégie and Menzies, 1980]. Considering an average ozone altitude distribution for mid-latitude regions [Krueger and Minzner, 1976] and the ozone absorption cross sections as given by Bass and Paur [1985], the optimum wavelength for measurements between 35 and 50 km at mid-latitudes for ground-based measurements is then 308 nm, corresponding exactly to the XeCl exciplex laser output wavelength.

The statistical error on the measurements can then be computed from the usual lidar equation, which relates the back-scattered signal  $P(\lambda, Z)$  at a given wavelength as a function of the altitude  $Z$ , to the lidar system parameters, and the atmospheric scattering process [Schotland, 1974]:

$$P(\lambda, Z) = \eta E(\lambda) \frac{A}{Z^2} [\beta_m(\lambda, Z) + \beta_p(\lambda, Z)] \cdot \exp - \{2(\tau_{O_3} + \tau_m + \tau_p)(\lambda, Z)\} \quad (1)$$

where  $\eta$  is the receiver optical efficiency;  $E(\lambda)$  the emitted energy at the wavelength  $\lambda$ ;  $A$  the telescope area;  $\beta_m$  and  $\beta_p$  the molecular and particular backscattering coefficients, respectively; and  $\tau_{O_3}$ ,  $\tau_m$ , and  $\tau_p$  the optical thicknesses due to ozone absorption, molecular, and particular extinction, respectively.

The accuracy  $\varepsilon$  on the measurements of the ozone number density  $n_{O_3}(Z)$  for a vertical resolution  $\Delta Z$  and a number of laser shots  $N$  is then given, in the absence of background noise, by

$$\varepsilon = \frac{1}{2\Delta\sigma_{O_3}(Z)n_{O_3}(Z)\Delta Z(N^{1/2})} \left[ \sum_{i,j=1}^2 \frac{1}{\bar{P}(\lambda_i, Z_j)} \right]^{1/2} \quad (2)$$

where  $\Delta\sigma_{O_3}(Z)$  is the differential ozone absorption cross section between the two emitted wavelengths  $\lambda_1$  and  $\lambda_2$ , and  $\bar{P}(\lambda_i, Z_j)$  is the backscattered signal averaged on  $N$  laser shots and filtered to obtain an altitude resolution  $\Delta Z$  at the wavelengths  $\lambda_i$  ( $i = 1, 2$ ) and for the altitudes  $Z_j = Z + (j - \frac{3}{2})\Delta Z$  ( $j = 1, 2$ ).

Such an expression will allow us to scale different systems or to perform a tradeoff between the relevant parameters of a given lidar system as it gives the dependence of the accuracy  $\varepsilon$

on the emitted laser energy  $E$ , the receiver efficiency  $\eta$ , the receiving telescope diameter  $D$ , the integration time  $t_i$ , and the height resolution  $\Delta Z$ ;

$$\varepsilon \propto t_i^{-1/2} \Delta Z^{-3/2} (E\eta)^{-1/2} D^{-1} \quad (3)$$

Taking into account the present status of development of exciplex lasers, the experimental results obtained in September 1983 have been extrapolated, assuming an on-line output energy of 250 mJ (20-Hz pulse repetition frequency) and an off-line output energy of 60 mJ (20-Hz pulse repetition frequency) for a respective receiver optical efficiency of 6 and 3% and a 0.5 m<sup>2</sup> area telescope. The results are shown on Figure 5 representing, as a function of altitude, the integration time required for a 2% accuracy on the measurement, considering nighttime conditions. Possible tradeoffs, as given by (3), will have also to be taken into account with respect to the scientific objectives of the measurements. Whereas long integration times, over a whole night, are possible for trend studies, shorter integration times are requested for studies of shorter-scale ozone fluctuations, leading to tentative correlations with temperature variations.

Besides the signal statistical errors, systematic errors of various types have also to be considered.

The absolute values of the ozone cross sections seem to be presently known in the Huggins bands below 320 nm with a  $\pm 3\%$  accuracy [Malicet et al., 1985]. However, the relative precision over the same spectral range is better, of the order of 1–2% [Paur and Bass, 1985]. Only the latter has to be considered here, as ozone number densities can always be recomputed from the lidar data, taking into account the latest determinations. Of particular importance is the temperature dependence of the cross sections in this wavelength range, which at 40 km, might introduce an error of about 0.2% per degree in the difference between the assumed and the true temperature profiles. However, using a combination of radiosonde and lidar data to derive the vertical temperature profile as mentioned previously, this error can be reduced to 0.5%, considering absolute temperature accuracies of about 1–2%.

Also to be considered is the influence of the laser line width and emitted wavelength fluctuations on the effective absorption cross-section determination. The natural emission width of the XeCl laser is of the order of  $\sim 0.1$  nm and corresponds to two to four different lines, depending on the operating pressure [Tellinghuisen et al., 1976]. In calculating the effective cross section, one has thus to take into account both the line positions and their relative intensity (cf. Figure 1) which should be checked during the experiment to maintain the systematic error below 0.2%.

The differential Rayleigh extinction for a 47-nm wavelength separation between the on (308 nm) and off (355 nm) wavelengths induces an error in the measured ozone concentration of the order of 3% at 40 km. It has thus to be accounted for, using either model average density profiles or radiosonde-calibrated lidar data. Such a correction will drop the associated systematic error below 0.1% for the altitude levels above 25 km.

Mie scattering due to aerosol particles also introduces a differential extinction error. Here again as for temperature and density corrections, one can use the lidar off-line wavelength to monitor the aerosol scattering ratios in the altitude range of the measurement. Under normal conditions the aerosol extinction coefficient above 30 km altitude is lower than  $2 \times 10^{-4}$  km<sup>-1</sup> [Elterman et al., 1969], which introduces a maximum

error of 0.5%, thus requiring no correction. If high-aerosol content is present, a correction should be made, assuming a knowledge of the nature and shape of the aerosol particles from other measurements and using aerosol models to reduce the error to a similar value. In this case a third wavelength in the UV range has to be used, as the atmospheric molecular scattering profile cannot be derived with a single off wavelength. It is important to point out here that due to the range-resolved nature of the lidar soundings, the measurements at a given level are insensitive to the aerosol content at lower altitudes.

Interferences due to absorptions by other minor constituents such as SO<sub>2</sub> and NO<sub>2</sub> also must be considered. Whereas such interferences are of importance for ozone measurements at very low altitudes [Pelon and Mégie, 1982a] the very low concentration of such species relative to ozone at altitude levels above 20 km reduces their values to less than 0.3% under normal conditions (NO<sub>2</sub> at 40 km), leading to errors lower than 0.1% after correction using an averaged profile.

When considering the overall uncertainty resulting from both statistical and systematic errors, as analyzed here, lidar measurements using up-to-date commercially available laser sources will have the potentiality to probe the ozone distribution in the upper stratosphere with a precision compatible with the geophysical objectives: at 40 km for a 4-hour integration time one can expect a global precision better than 3% for a 3.5-km altitude resolution, whereas the same precision will require an integration time of 6 hours and an altitude resolution of 7 km at 45 km.

#### CONCLUSION

The use of powerful exciplex laser sources directly emitting in the UV spectral range has allowed lidar monitoring of the ozone vertical distribution up to the 48-km altitude level. Extrapolations show that precisions better than 3% are within the range of the expected performances of updated lidar systems. Such a station will be implemented at the Observatoire de Haute Provence to determine, from a statistical analysis of experimental uncertainties and ozone short-term variations, the potentiality of the lidar system for network monitoring of ozone trends in the upper stratosphere.

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